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PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Matthias KRULL et al.

Docket: 2002DE444

Serial No.: 10/735,490

Group Art Unit: 1714

Filed: December 12, 2003

Examiner: TOOMER, C.

For: FUEL OILS HAVING IMPROVED COLD FLOW PROPERTIES

DECLARATION UNDER 37 CFR 1.132

Mail Stop Amendment
Commissioner for Patents
PO Box 1450
Alexandria, VA 22313-1450

Dear Sir:

I, Matthias Krull, state that I am a resident of Am Rheinhessenblick 27, D-55296 Harxheim, Federal Republic of Germany; that I am a citizen of the Federal Republic of Germany; that I am a chemist having earned the degree of Dr. rer. nat. (corresponds to Ph. D.) from the Free University Berlin, Federal Republic of Germany, in 1989.

I am acquainted with the subject matter of the above subject Application and I am one of the named inventors of Application No. 10/735,490, filing date of December 12, 2003 in the name of Matthias Krull et al. for "FUEL OILS HAVING IMPROVED FLOW PROPERTIES."

I have been employed for 17 years in the Research and Development department of Hoechst AG, Frankfurt, Germany, which was succeeded by Clariant GmbH, Frankfurt, Germany, where my work has focused on oilfield chemicals and especially on cold flow additives for mineral oils.

CERTIFICATE OF MAILING/TRANSMISSION (37 CFR 1.8a) and 1.10

I hereby certify that this correspondence is, on the date shown below, being transmitted by facsimile to the U.S. Patent and Trademark Office. (Fax No. (571) 273-8300 (4 pages)

Jackie L. Wise:

Date:

Jackie L. Wise
Feb. 12, 2008

For comparison of the additive mixtures of Brown (US Patent No. 5,906,663), herein after referred to as the Brown Patent, with terpolymers according to the invention, I prepared the following CFPP and filterability tests in a low sulfur, low aromatics fuel oil (Test oil 5). The characterization of the test fuel oil was made according to the methods given in the subject application as filed.

	Test oil 5
Distillation	
IBP [°C]	176
20 % [°C]	235
90 % [°C]	328
FBP [°C]	348
Cloud Point [°C]	-9,9
CFPP [°C]	-11
Paraffin 10°C below CP (DSC)	3,7 %
Density@15°C [g/cm³]	0.834
Sulfur content [ppm]	9
Aromatic content [% by weight]	17,6
of which mono [% by weight]	16,7
di [% by weight]	0,9
poly [% by weight]	<0,1

In this Test oil, additives P1, P6 and P8 as characterized in Table 1 of the subject application (containing 65% active in kerosene) were compared with the additive combination as exemplified in the Brown Patent. In the Brown Patent, terpolymer A is said to be Dodiflow-V-4159, which was sold commercially by Hoechst. Clariant, as the legal successor of Hoechst AG in this technical field, is in the position to say that this Dodiflow-V-4159 contained approximately 16 mol-% vinyl acetate and approximately 1.2 mol-% vinyl ester of neodecanoic acid. A similar product was already cited as Comparative Example P14 in the subject application. It was shown that this additive

has only borderline solubility in fuels with low sulfur and low aromatics. Here Dodiflow-V-4159 was used as comparative additive P"A".

The Brown Patent discloses that in addition to the "A" component, an additive component B which is an ethylene-vinyl acetate copolymer is required. For comparison purposes in the following examples used a component P"B" which was a very similar material to component B of the Brown Patent, which was an ethylene-vinyl acetate polymer with a number average molecular weight of 5200 (GPC) and a vinyl acetate content of 13.7 wt.-% (equivalent to 4.9 mol.-%) copolymer (P"B").

CFPP effectiveness in test oil 5

Example No.	Additive	300 ppm	500 ppm	700 ppm
62	P1	-18 °C	-20 °C	-22 °C
63	P6	-19 °C	-21 °C	-24 °C
64	P8	-19 °C	-22 °C	-25 °C
65 (comp.)	P"A" + 2% P"B"	-16 °C	-18 °C	-19 °C
66 (comp.)	P"A" + 5% P"B"	-18 °C	-20 °C	-21 °C

To test the solubility of the terpolymers according to the invention in comparison to the additive mixtures of the Brown Patent, 500 ml of Test oil 5 were admixed at 25°C with 500 ppm of the additive, respectively to provide an additized oil sample. All of the additive mixtures to be tested were admixed at 25 C, and all additives were a 65% active concentration in kerosene. The solubility test was performed according to the description in the subject application. The results of the solubility determination are shown in the following table which presents data representing the filterability of the Test Oil/Additive mixture. Filterability is determined by the time required to filter the additized test oil sample. Filter times in excess of 120 seconds are considered unacceptable.

Filterability of Additized Test Oil 5

Example No.	Additive	oil temperature	Time [sec]	Volume [ml]
67 (comp.)	none	25 °C	55	500
68	P1	25 °C	63	500
69	P6	25 °C	59	500
70	P8	25 °C	60	500
71 (comp.)	P14 + 2% P"B"	25 °C	>120	approx. 450
72 (comp.)	P14 + 5% P"B"	25 °C	>120	approx. 300

These experiments clearly show that the terpolymers of the invention containing structural units derived from vinyl acetate and units derived from a vinyl ester of a tertiary branched carboxylic acid in the specified molar ranges have excellent solubility in middle distillates having low sulfur and aromatics content, even at low blending temperatures. In contrast to the additives of the subject invention, the additive combinations of the Brown Patent resulted in filter blockages (i.e., they have unacceptable filtration times >120 sec). Especially at higher contents of component B, which are necessary for an improved CFPP performance, the additive combinations of the Brown Patent are not fully soluble under cold blending conditions. The lack of solubility at cold blending conditions potentially will result in undesired filter blockages. Furthermore, the terpolymers of the subject invention show an improved CFPP performance over Brown. The terpolymers of the instant invention provide economic advantage in the adjustment to the CFPP of the fuel oils with lower dosage rates than are required by the additive mixtures of Brown for the same fuel oil.

The terpolymers of the invention are equally suitable for improving the cold flow properties of mixtures of fatty acid methyl esters with mineral diesel fuel. This is shown by CFPP measurements in Test oil 5 (characterization shown hereinabove) also containing varying amounts of rape seed methyl ester. The rape seed methyl ester used comprises 62,2% oleic acid methyl ester, 19,7 % linolic acid methyl ester, 8.9% linoleic acid methyl ester, 4,6 % palmitic acid methyl ester and 1,5 % stearic acid

methyl ester as the main components. The following table shows the impact of additives of the instant invention on the Cold Filter Plugging Point (CFPP) of biodiesel mixtures based on Test oil 5 and varying amounts (5, 15, and 25 %) of rape seed methyl ester.

CFPP Effectiveness in mixtures of Test oil 5 with Varying Amounts of Rape Seed Methyl Ester (RME)

Example no.	test oil 5	RME	additive	200 ppm	400 ppm	700 ppm
73	85 %	15 %	P1	-16 °C	-19 °C	-21 °C
74	85 %	15 %	P6	-15 °C	-20 °C	-23 °C
75	85 %	15 %	P8	-17 °C	-20 °C	-22 °C
76	95 %	5 %	P6	-17 °C	-21 °C	-24 °C
77	75 %	25 %	P6	-14 °C	-17 °C	-21 °C

Usually the addition of fatty acid methyl esters improves the solubility characteristics of mineral diesel blends towards cold flow additives. Nevertheless, also in such mixtures of mineral diesel fuel with fatty acid methyl esters, the additive mixtures of Brown especially upon cold blending conditions lead to fuels having poor filterability. In contrast, the terpolymers of the instant application under the same blending conditions give fuels of superior filterability for fuels having from 5 to 25 % fatty acid methyl esters as shown below (test conditions equal to above those described above).

Filterability of additized mixtures of test oil 5 with Varying Amounts of Rape Seed
Methyl Ester (RME)

Example No.	test oil 5	RME	Additive	Oil temperature	Time [sec]	Volume [ml]
78 (comp.)	85 %	15 %	none	25 °C	48	500
79	85 %	15 %	P1	25 °C	56	500
80	85 %	15 %	P6	25 °C	50	500
81	85 %	15 %	P8	25 °C	51	500
82 (comp.)	85 %	15 %	P14 + 2% P"B"	25 °C	>120	approx. 470
83 (comp.)	85 %	15 %	P14 + 5% P"B"	25 °C	>120	approx. 340
84	95 %	5 %	P6	25 °C	55	500
85	75 %	25 %	P6	25 °C	47	500
86 (comp.)	75 %	25 %	P14 + 5% P"B"	25 °C	>120	approx. 380

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Frankfurt am Main,

Date: 08.02.2008
Matthias Krull